University of
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DETERMINATION OF RADIUM-226 IN URANIUM BEARING REFERENCE MATERIALS FOR CERTIFICATION PURPOSES

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Determination of Radium-226 in uranium bearing reference materials for certification purposes

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Four uranium bearing reference materials BL-4a, DH-1a BL-5 and DL-1a received from CANMET were analyzed for Ra-226 content. Radium was co-precipitated with barium and lead as sulfate. Lead and thorium was removed by dissolving the precipitate in diethylenetriaminepentaacetic acid and re-precipitating barium sulfate with 20 per cent Na_2SO_4 in the presence of acetic acid. The precipitate was deposited on a solid state detector and a pulse height analyzer. Events due to particle from ^{226}Ra were integrated for the estimation of ^{226}Ra. The chemical recovery was monitored by adding ^{133}Ba along with barium carrier.

Each sample was dried in an oven at 110°C and then weighed with an accuracy of 20 microgram. The weighed sample was transferred to a platinum crucible for performing fusion with potassium pyrosulfate. The sample was heated for ashing all organic material. 3 ml of H_2SO_4 and 15 ml of HF was added and the mixture heated to ensure no silica remained in the sample. 5 g of potassium pyrosulfate was added and the sample was cooled and dissolved in 10% HCl and made to a known volume.

An aliquot of solution or the whole solution was then

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taken for ^{226}Ra analysis. An appropriate amount of 50% H_2SO_4 and K_2SO_4 was added to the solution and then heated to near boiling. ^{133}Ba, $BaCl_2$ and $Pb(NO_3)_2$ solutions were added to the sample while it was being stirred. The stirring was continued for thirty minutes. The precipitate was removed by centrifugation and then dissolved in 0.17 M DTPA. Any undissolved material was removed by centrifugation and Na_2SO_4 solution was added and mixed thoroughly. The sample was heated for 5 minutes after the addition of 2ml of 6M acetic acid. $BaSO_4$ was removed by centrifugation and the dissolution by DTPA and precipitation cycle was repeated. The precipitate was then deposited on a special filter paper for α spectrometry. The filter was dried and placed on a planchet and assayed for α activity by using a solid-state detector and a pulse height analyzer. A scintillation counter was used for γ-ray assay.

The geometry factor was determined by using a standard Ra-226 solution. The radium solution was obtained from the U.S. National Bureau of Standards. A copy of the certificate and dilution procedure is enclosed herewith as Appendix I. The activity of the solution was 1966.7 $\pm$ 29.8 pCi/ml. The geometry factor was also determined by using standard ^{241}Am source.

The α and γ counts along with weight of each sample are given in Table I, along with one standard deviation evaluated on the basis of counting statistics.

It appears that DH-1a contains barium and therefore relatively 'thicker' samples were used for α spectrometry. The α particle peaks in the spectrum had long tails which required estimation of overlap of α peaks due to other emitter. However, soon after precipitation the samples were counted so that substraction of events from other particle peaks was not large. The agreement among the replicates was found to be within 5%.

TABLE I.

Reference	Weight (g)	Counts (ct/sec)	Counts (ct/min)	Activity (pci/g)	Standard deviation %
BL-4a	1.000	21349/440905	56245/90 min	388.5	0.80
	1.000	64370/328163	123390/90 min	395.9	0.48
	1.008	19589/140770	63100/40 min	399.6	0.82
	1.041	19762/34882	75010/20 min	418.3	0.80
DH-1a	1.350	40829/38463	71005/30 min	833.9	0.62
	1.000	80433/55139	52720/20 min	845.6	0.56
	1.000	15875/18911	52506/10 min	842.2	0.90
	0.8097	20228/97438	60275/60 min	850.0	0.81
BL-5	1.00105	17754/38394	59840/20 min	23,500	0.85
	1.00230	42458/30749	76066/10 min	24,016	0.61
	1.00230	14755/15343	74911/10 min	24,205	0.90
	1.00105	16693/24359	71415/20 min	23,202	0.86
DL-1a	2.000	19189/232012	53020/20 min	37.2	0.84
	2.008	17090/86866	56090/10 min	35.76	0.87
	2.502	34611/77933	55715/10 min	36.4	0.68
	2.500	61972/144933	52410/5 min	36.3	0.59

Radium-226 Reference Solution

Prepared 30 June 1982 from NBS SRM 4959-158 by J.L. Dalton

Contents of the ampoule were transferred quantitatively to a 1 l volumetric flask (with multiple rinses of the ampoule with water). Five ml of concentrated hydrochloric acid were added, and the solution was diluted to the mark and mixed thoroughly.

Calculation of Present activity (Confirm to your own satisfaction)

Time since certification: 15.3 y

Half-life of radium-226 : 1600 y (Int.J.App.Rad.Isotopes,2,141(1959) Nukleonik 8,244(1966)Chem.Ant.65, 6599 (1966)

$$\ln \frac{A_t}{A_o} = \ln \frac{N_t}{N_o} \cdot \frac{t}{t_{½}} = -.006628$$

$$\therefore \frac{A_{t\ present}}{A_o} = 0.9934$$

Specific activity of ^{226}Ra $(3.659 \pm .015) \cdot 10^{10}$ Bq/g $(\pm .38)$ (H.Ramthun, C.a.65,6599 (1966))

Consequently, calculated activity of solutaion is

$A = 2.002 \times 10^{-6} \times 0.9934 \times 3.659 \cdot 10^{10}$ Bql^{-1}

or $A = 7.277 \cdot 10^{4}$ Bql^{-1}

= 72.77 Bq/ml ($\pm$max.1.10 Bq/1ml) ($\pm 1\sigma$ random 0.20 Bq/m/)

= 1966.7 pc:/ml ($\pm$max 29.8 pCi/m/) ($\pm 1\sigma$ random 5.3 pCi/m/)

Calculation of uncertainty in activity

1 relative error in $t_{½}$: $\sim \frac{2.3}{1600} = \sigma_A$

1 relative error in sp. activity: $\frac{5}{3659} = \sigma_B$

1 relative error in µg radium: $0.0018 = \sigma_C$

Combined random statistical error: $\sigma_D = (\sigma_A^2 + \sigma_B^2 + \sigma_C^2)^{½} = 0.0027 = 0.27\%$

Systematic uncertainty in dilution (estimated)	0.02%
Systematic uncertainty in μg Ra	0.80%
	0.82%

Maximum conceivable error (99% conf.) = 2.58x0.27%+0.82%

= 1.52%

U. S. Department of Commerce
Alexander B. Trowbridge,
Secretary

National Bureau of Standards
A. V. Astin, Director

Certificate
Standard Reference Material 4959
Gamma-Ray Standard
Radium—226

This standard consists of radium 226 in approximately 5 milliliters of solution in a flame sealed glass ampoule. The solution is 5 percent by weight HNO_3.

The weight, in micrograms, of radium 226 contained in the ampoule as of September 1, 1967, was

$$A \pm 2_c \pm 2_{sc}$$

This standard was calibrated by comparing its gamma ray emission rate with those of a 1957 series of standards prepared from material that had been compared in the National Bureau of Standards radium balance with the national radium standards. The gamma ray emission rates were compared in a $4\pi\gamma$ ionization chamber containing argon at a pressure of 10 atmospheres.

The uncertainty, 2_c percent, is the sum of 0.1_6 percent, which is the statistical error at the 99 percent confidence level ($2.58\ s_m$, where s_m is the standard error), and 0.8_0 percent, which is the maximum uncertainty due to assessable systematic errors in the 1957 series of standards and in the ionization chamber comparisons.

This standard was calibrated in the Center for Radiation Research by members of the Radioactivity Section, W. B. Mann, Chief.

Washington, D. C. 20234
March 1, 1968

W. Wayne Meinke, Chief
Office of Standard Reference Materials

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